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Nonlinear optical laser devices

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Abstract : Phase-matched devices based on nonlinear three-wave interaction of laser radiation in crystalline medium are discussed. These include efficient second harmonic generator, tunable parametric oscillator (OPO), tunable infrared generation by difference-mixing and infrared detection by parametric up-conversion. The nonlinear crystal forms the cornerstone of such devices. After explaining the requirements on crystal properties the method of improving and enhancing the nonlinear effect is discussed for efficient operation of the device. Emphasis is made on the tunable infrared generation and the characteristic features are pointed out.

1. Introduction

In a linear system the response is proportional to the external influence. All transparent materials were regarded as linear systems in optics since the time of Newton who recognised noninteraction of light beams in a material medium. It came to reality only with the advent of lasers, whose intense optical electric field is capable of driving the valence electrons sufficiently strong enough to the anharmonic regime and yielding thereby measurable nonlinear effects. The aim of the paper is to introduce nonlinear devices arising out of the physical phenomena. Our discussion is limited to only nonlinear devices steming from the second-order polarisation in three-wave interactions. The four-wave interaction in gases and vapours arising out of third-order polarisability is recently gathering momentum and can also under certain conditions give rise to devices similar to the three-wave process. Out of the variety of nonlinear interactions (Bloembergen 1978) we however prefer to concentrate our attention to the much well-developed and well-established techniques. The basic nonlinear processes and the means for their enhancement is considered in section 2 and section 3 gives the present state-of-the-art of some practicable nonlinear devices.

2. Nonlinear processes

2.1 *Interactions*

The dielectric polarisation P induced by the application of optical electric fields E, is in general given by

$$P_i = \chi_{ij}E_j + \chi_{ijk}E_jE_k + \chi_{ijkl}E_jE_kE_l + \cdots$$

where χ's denote the susceptibility tensors of different ranks whose magnitude rapidly decreases for higher orders and consequently higher laser power is required to observe noticeable effect. The first or the lowest-order nonlinear term is quadratic in electric field which leads to optical rectification and second harmonic generation (SHG), sum and difference mixings, and parametric amplification/oscillation (OPO). The third term which is the lowest-order nonlinearity in centrosymmetric media such as gas, vapour, can give rise to several measurable effects like third harmonic generation, Kerr effect, electric field-induced SHG, nonlinear refractive index, two-photon absorption, different four-wave mixings and so on.

2.2 *Aspects of phase matching*

Like any other phenomena, the nonlinear processes are required to satisfy conservation laws. While the process is identified by the energy conservation, the conservation of momentum is demanded by the necessity of the interaction to constructively add-up throughout the entire crystal length (not done in original experiment Franken *et al* 1961) and thus is done through correct adjustment of phase velocity amongst the incident and generated beams. The deleterious effect of the crystal normal dispersion in this matter is overcome by a proper choice of polarisation and propagation direction of the interacting waves in the nonlinear crystal taking it to be anisotropic. The consideration leads to a term $(\mathrm{Sin}\ X/X)^2$, where $X = \Delta kl/2$, l is the crystal length, and Δk is the wave-vector mismatch.

Two effects of phase matching, though interrelated need particular mention in connection with devices . achievement of noncriticallity in phase matching and angular or acceptance bandwidth. The dependence on other factors has been illustrated in figure 1. When phase matching occurs in a direction other than 0° or 90° to the optic axes of the crystal, the physical separation between the interacting beams through double refraction leads to a limit to the usable crystal length. This is overcome by adjusting, if possible, the phase matching to 90° called the noncritical matching. This is addition provides a large acceptance angle, a situation highly desirable for efficient device operation. Such a non-critical situation may be realised using temperature dependency on refractive indices (Koechner 1976) and also by introduction of noncollinearity in phase matching. The later property in addition is capable of phase matching simultaneously a broad band-width signal, a situation particularly desirable for infrared up-conversion (Zerniko and Midwinter 1973).

2.3 *Desirable crystal properties*

Apart from the basic need of sufficient crystal birefringence for phase matching, it is necessary that all the interacting waves should be within the fundamental optical transmission range. The short wavelength limit is set by the

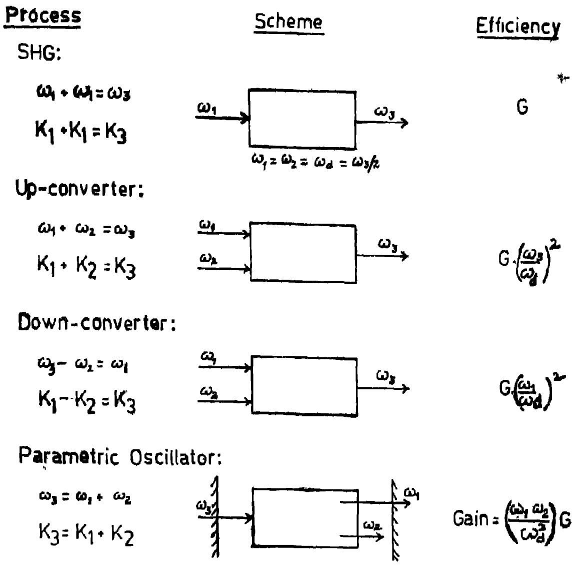

Figure 1. Three-wave frequency mixing processes. G represents the gain

$$G = \frac{2\omega_d^2 d^2 l^2 I}{n_1 n_2 n_3 c^3 \epsilon_0}$$

where d is the effective nonlinear coefficient I is the pump intensity, n's are the refractive indices at the frequencies of the interacting waves. ω_d is the degenerate frequency.

band-edge while the long wavelength limit may be taken to be that imposed by two-phonon lattice vibration (Bhar and Smith 1972). The incorporation of light elements in the crystal reduces the longwavelength limit (Table 1). Besides atomic masses, force-constant also considerably influence (Bhar 1978). The additional absorption features within this range include that due to impurity, intra-valence band, free-carrier, three-phonon, stoichiometric defect, and these can affect device operation in many ways. For example it reduces conversion efficiency, accelerates crystal damage threshold and raises oscillation threshold of OPO. The extrinsic absorption can be removed through refined crystal growth technique and special heat treatment. In this way considerable improvement in crystal quality (figure 2) has been effected in $AgGaS_2$ for example (Bhar and Smith 1974, Mathes *et al* 1975, Route *et al* 1976). The laser damage in addition is associated with avalanche ionisation resulting from self-focussing of the incident laser beam within the crystal (Bloembergen 1974)

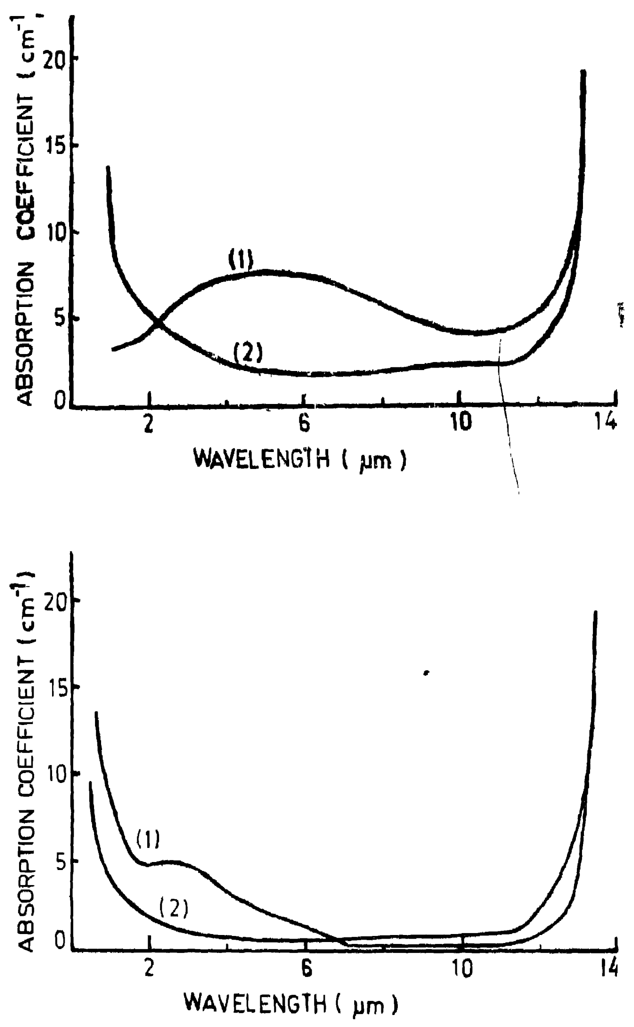

Figure 2. Removal of residual absorption in $AgGaS_2$ by annealing in vacuum Upper figuro : (1) before, and (2) after annealing an yellow sample. Lower figure : (1) before and (2) after annealing a milky sample.

One more important aspect determining conversion efficiency in nonlinear devices is the nonlinear figure of merit which is given as d^2/n^3 (figure 1), d is the effective nonlinear coupling coefficient and n the refractive index. A high value of nonlinearity is therefore an obvious requirement Attributing the intrinsic nonlinearity to bond ionicity and asymmetry in atomic sizes, Levine (1973) was able to successfully account for nonlinearity in a wide variety nonlinear crystals and identified crystals of high nonlinearity. Nonlinear properties of such crystals have been reviewed (Smith 1975 and Iseler 1977).

Table 1. Properties of some established nonlinear crystals.

Crystal (Point group)	n_{av}	B	Transmission Range (μm)	Nonlinear Coeff. $d\times10^{12}$ (m/v)	Damage threshold MW/cm^2
KDP ($\bar{4}$ 2 m)	1·5	−·04	·2—1·1	·5	1000
$LiNbO_3$ (3 m)	2 2	−·08	·3—4 5	6·2	50
Ag_3AsS_3 (3 m)	2·6	−·22	·6—13	11·6	20
CdSe (6 mm)	2·4	·02	7—25	19·0	60
GaSe ($\bar{6}$ m 2)	2·7	−·35	·6—18	88·5	30
$AgGaS_2$ ($\bar{4}$ 2 m)	2·4	−·05	·6—13	12·0	20
$AgGaSe_2$ ($\bar{4}$ 2 m)	2·6	−·03	·7—18	33·0	11
$ZnGeP_2$ ($\bar{4}$ 2 m)	3·1	·04	·7—12	75·0	>4
$CdGeAs_2$ ($\bar{4}$ 2 m)	3·6	·09	2·4—18	236·0	38

2.4 *Nonlinear crystals*

The established crystals listed in table 1 can be classified according to their transmission limits. UV transmitting crystals are those belonging to KDP family which includes Ammonium, Rubydium and Cesium in place of K, Arsenate in place of P and the deuterated versions. These dielectric crystals transmit up to about 1·5 μm covering the visible The nonlinearity is rather low Ferroelectric crystals like $LiNbO_3$, BSN transmit further into the infrared 5 μm and possess higher nonlinearity. Similar is the case for $LiIO_3$. The high nonlinearity is exhibited by the infrared transmitting crystals and these are minerals like Ag_3AsS_3, Ag_3SbS_3 and Tl_3AsSe_3; semiconductors like CdSe, GaSe, $ZnGeP_2$, $CdGeAs_2$, $AgGaS_2$ and $AgGaSe_2$ Some of these show only marginal visible transmission which are particularly suited for nonlinear experiments using dye lasers. The specific applications of the crystals are discussed in the following section.

3. Features of nonlinear devices

The present state-of-the-art of the following three categories of nonlinear devices are discussed : second harmonic generator, infrared detection by up-conversion and tunable lasers based on down-conversion and optical parametric oscillation.

3.1 *SHG*

SHG is the first nonlinear effect to be studied in any nonlinear crystal. It gives practicable methods for the measurement of nonlinear coefficient (Kurtz 1975) and determination of phase matching behaviour (Bhar and Smith 1974) and hence helps in the assessment of nonlinear crystals. It can provide convenient pump wavelengths for OPO when the fundamental laser wavelength does not permit favourable tuning range. For example, ruby laser SH pumped OPO in $LiIO_3$ provides tuning range 0·41–2·1 μm (Nath and Pauli 1973); fourth harmonic of Nd laser pumped OPO in ADP provides the tuning of the entire visible range (Yarborough and Massey 1971). As it has been pointed out, SHG requires the maximum birefringence for phase matching compared to other interactions and so absence of SHG in CdSe does not preclude it for other mixing processes. Like any other nonlinear interaction, the efficiency of conversion in SHG can be improved by judiciously selecting noncritical phase matching angle. This has been possible in ferroelectric crystals like $LiNbO_3$, BSN and also in KDP type of materials by using temperature variation of refractive indices. Adhav and Vlassopoulos (1974) have given temperature tuned wavelengths for 90° matching in KDP isomorphs. The introduction of noncollinearity in phase matching is another possibility. Conversion efficiencies more than 50% were obtained. Geusic have obtained 100% conversion efficiency for intracavity SHG in BSN by a Nd laser. Efficiency is low for infrared SHG, Iseler *et al* (1977) reported 27% energy conversion efficiency in $CdGeAs_2$ by CO_2 laser.

SHG is also used as frequency multipliers. The efficient frequency doublers using $LiNbO_3$, $LiIO_3$ or BSN are unique power sources of coherent radiation. Doubling of tunable dye lasers in KDP isomorphs provides efficient tunable sources in the UV.

3.2 *Up-Converter*

Up conversion is a special case of sum frequency generation in which a short wavelength pump laser radiation is added with a long wavelength infrared radiation in a nonlinear crystal. This parametric process can be used to shift the frequency (and hence the information contained thereby) of an infrared beam to the visible region by adding up with an intense visible-near-infrared laser beam thereby making available sensitive detectors in the visible for the detection of weak infrared radiation. Thus this technique not only provides an effective fast and sensitive infrared detector but also offers the possibility of operating the detector at room temperature. The competitive detectors for the infrared, on the other hand usually operate at a low temperature. The technique has also fruitfully been utilised for the detection of thermal radiation from astronomical objects (Abbas *et al* 1976).

The subject of up-conversion for the detection of infrared signal and image has been reviewed by Warner (1971), and Zernike and Midwinter (1973). Detailed system studies have also been carried out by these workers The two basic design parameters for an up-converter are · field-of-view and acceptance angle Both are determined by the phase matching angle and is large for 90° phase matching. Apart from large field-of-view and acceptance angle, simultaneous phase matching of a large band-width signal is also desirable. Most work on up-converter was done by Warner and Midwinter in proustite and $LiNbO_3$ The latter though provides noncritical matching but can not be used beyond say, 4 μm. Because of the large nonlinear figure of merit and the possibility of noncritical phase-matching, the chalcopyrite $ZnGeP_2$, $AgGaS_2$, and $AgGaSe_2$ crystals have been shown to be very promising (Bhar 1974) and experiments have subsequently been reported for up-conversion of 10·6 μm radiation in such crystals (Jantz and Koidl 1977, Andreeva *et al* 1979)

3.3 *Optical Parametric Oscillator*

The spontaneous decay of a pump photon in a nonlinear crystal into two photons (signal and idler) is known as parametric fluorescence Enhancement of the generated photons can take place under the usual phase matching condition and also by resonance in an optical cavity. When the gain exceeds the cavity loss the device reaches the threshold for oscillation and the output power at signal/idler frequency comes out through the partially transmitting resonator mirror. Because of large parametric decay probability the gain is maximum at the degenerate frequency. Frequency tuning i.e. oscillation at off-degenerate frequency can be obtained by altering the phase matching condition through a variation of refractive indices. The later may be effected by a change in crystal orientation, temperature.

The characteristics of an OPO are · threshold, tuning, line-width and conversion efficiency (Byer and Herbst 1977) The crystals must be of very high quality otherwise the pump laser may damage the crystal before reaching the OPO threshold. Conversion efficiency as high as 45% has been obtained in Nd laser pumped OPO in $LiNbO_3$. Tuning in the visible though obtained with

KDP, ADP, $LiNbO_3$, and BSN is not that important because of the availability of tunable dye lasers in the region. OPOs developed to-date provide tuning from the visible to 10 μm. Range of tuning obtainable is determined by the location of the pump which should lie near the shorter wavelength transmission limit of the crystal for wide tuning In the near infrared region tuning has been obtained in a number of crystals e.g. BSN, $LiNbO_3$, $LiIO_3$. For the medium infrared region only CdSe and Ag_3AsS_3 have so far been used for OPO. Ternary crystals (table 1) provide low oscillation threshold and wide tuning range, but lack of good quality crystals has prevented further development Table 2 represents some OPOs covering important tuning ranges. The line-width of the output is $\sim$ cm^{-1} but can be reduced considerably by using a Fabry-Perot etalon and or grating in the cavity. Tuning for ferroelectric crystals can be made both through angle and temperature variations but for other through angular tuning only

Table 2. Wide tunable infrared generation by some nonlinear optical devices

Device	Non-linear crystal	Pump laser/s	Tuning range (μm)	Reference
OPO	$LiNbO_3$	Nd : YAG	1·4–4·4	Herbst *et al* 1974 *Appl. Phys. Lett.* 25 520
OPO	Ag_3AsS_3	Nd : $CaWO_4$	1·2–8·5	Hanna *et al* 1973 *Appl. Phys. Lett.* 22 440
DC	Ag_3AsS_3	Dye and Dye	11–23	Hocker and Dewey 1976 *Appl Phys.* 11 137
DC	$AgGaS_2$	Dye and Dye	5·5–18·3	Seymour and Zernike 1976 *Appl Phys. Letts.* 29 705
DC	$AgGaS_2$	Ruby and Dye	4·6–12	Hanna *et al* 1973 *Opts. Comm.* 8 151
DC	GaSe	Ruby and Dye	9 5–18	Abdulaev *et al* 1976 *Sov J. Quan. Elect.* 6 88
DC	GaSe	YAG and Dye	9–19	Oudar *et al* 1979 *Opts. Commun.* 29 119
OPO-DC	Ag_3AsS_3	Proustite OPO	8–12	Bhar *et al* 1972 *Opts. Comm.* 6 323
OPO-DC	CdSe	Proustite OPO	9·5–24	Hanna *et al* 1974 *Appl. Phys. Lett.* 25 142
OPO-DC	$AgGaSe_2$ GaSe	$LiNbO_3$ OPO	4–18	Bianchi and Garbi 1979 *Opts Commun.* 30 122

3.4 *Difference-Mixers*

Two fixed frequency inputs provide only a fixed output difference-frequency; but if one of the input frequencies is varied a tunable output will be obtained As the output lies on the low frequency side this provides a method of generating tunable infrared radiation. In view of technological problems involved in operating an OPO in the medium infrared region, such a method is being increasingly used for the generation of tunable infrared radiation. The following three types of difference-mixer are considered : Down-converter (DC), OPO-DC and Backward-wave mixer. The backward-wave mixing is not discussed since not much study has been made. So far only one experiment (Cotter *et al* 1974) has been reported. Boyd *et al* (1972) have considered $ZnGeP_2$ for the generation of far infrared radiation.

3.4.1 *Down-Converter*

In a regular down-converter one of the high frequency lasers is taken to be a dye laser whose frequency is tunable. A small tuning in the dye laser appears as relatively large wavelength tuning in the infrared. The other pump laser is commonly a ruby system. However, a second dye laser has also been used. For CW operation argon ion laser is often used. The nonlinear crystal must be transmitting both at the input and the generated frequencies. And the provision to use a dye laser limits its use to only visible transmitting nonlinear crystals. Two types of crystals are used. In one type of crystals like $LiNbO_3$, $LiIO_3$, near infrared radiation is generated while in the other type Ag_3AsS_3, Ag_3SbS_3, $AgGaS_2$ medium infrared radiation is generated. Using GaSe oudar *et al* (1979) have generated infrared radiation upto 17 μm by mixing Nd laser and the same pumped dye laser radiations. The line-width of the output is limited to the line-width of the input pump lasers. The output power depends on the input pump power levels and powers from few watts to several Kilowatts have been obtained. Table 2 lists some important down-converters covering near and medium infrared regions. Tuning on the short-wavelength side is usually limited by the crystal birefringence while on the long wavelength side by the transmission cut-off of the crystal. And it is effected by simultaneous tuning of the dye laser and adjusting the crystal to the new phase matching angle.

3.4.2 *OPO-DC*

In this scheme the two outputs (viz. the signal and idler) of an OPO are difference-mixed in a second nonlinear crystal. The limitations of OPO on extension to infrared tunability and DC on phase-matched visible transmitting crystal are simultaneously overcome in this composite device in the following manner. OPO being a resonant device demands a very high quality crystal while the DC does not. The extension of tuning range in an OPO is beset with technological problem of design of a corresponding broad-band mirror coating. If however the outputs from a limited tunable OPO are difference-mixed in a second crystal which can tolerate poor crystal quality, a small OPO tuning results in a large wavelength tuning in the infrared, and at the same time the problem of obtaining a crystal with simultaneous visible transmission (since dye lasers are only available in the visible for DC experiments of 3.4.1) and wide transmission into the infrared is avoided. The DC crystal is only needed to start transmission near the degenerate frequency of the OPO.

This novel technique was first proposed and demonstrated (Bhar *et al* 1972) using a proustite OPO in a second proustite crystal. Nd laser pumped OPO tuning of 1·87–2·47 μm yields an output tuning 7·8–11·9 μm. The feasibility of the new technique has since been proved in many other nonlinear crystals and tunable radiations in the infrared have been generated, where no other technique could produce such a wide range. The possibility in CdSe originally (Bhar *et al* 1972) predicted for a tuning range 10–25 μm has since been demonstrated by Hanna *et al* (1974) and Andreou (1978) Using a $LiNbO_3$ OPO Bianchi and Garbi (1978, 1979) have covered the range 4–18 μm in GaSe and $AgGaSe_2$. The available tuning range in OPO-DC is summarised in table 2.

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